The spectroscopic foundation of radiative forcing of climate by carbon dioxide

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Outline

- Historical background of L-B-L calculation
- Motivation for radiative forcing research
- Definition and Examples of Radiative Forcing (RF)
- Uncertainty in RF due to Spectroscopic Parameters
  - Line shape and line mixing (Focus)
  - Line Strengths
  - Air-broadened halfwidths
- Results
- Utility of the Voigt function
- Summary and Conclusions

Line-by-Line Radiative Transfer Calculation

- **LBL calculation, a principal tool today, began in earnest at U. Michigan under Prof. S. Roland Drayson**

- **Drayson, a mathematician and an atmospheric scientist, began doing LBL calculations in the mid-1960's**

- **This 2016 is the 50th anniversary of his landmark paper on LBL calculation in Applied Optics**

- **Drayson computed transmittances for many of the early satellite experiments and was principally responsible for the spectral databases and transmittances for the LIMS experiment**

- **And, relevant to this talk, Drayson foresaw sub-Lorentizian wings in CO_2, noting in his *Appl Opt* paper that the Voigt profile would not be sufficient in the far wings of CO_2 bands!**

Atmospheric Transmission in the CO_2 Bands Between 12 μ and 18 μ

S. Roland Drayson

Calculations have been made of high-resolution transmission in the CO_2 absorption bands between 12 μ and 18 μ by direct integration across the bands, for both homogeneous and atmospheric slant paths. Mixed Doppler-Lorentz broadening has been used at pressures lower than 100 mb. A method to eliminate the Curtis-Godson approximation has been developed and applied to the slant-path calculations. Comparisons have been made with previous theoretical and experimental data, and reasons for the discrepancies are discussed.

Introduction

In recent years there has been a growing interest in atmospheric infrared radiative transfer, with applications to the earth and other planets. On the earth, these applications include the investigation of radiative heating and cooling, and the interpretation of satellite radiometer measurements, while the composition, surface pressure, temperature, etc., of planetary atmospheres may be inferred from suitable remote radiometric observations. Broad-band radiometers are being supplemented and replaced by instruments of much higher spectral resolution and photometric accuracy, leading to increased demands on the accuracy of calculations.

One of the chief problems in the interpretation of data stems from the difficulty in calculating atmospheric transmission functions due to molecular band absorption. These functions are generally obtained in one of two ways.

(a) From laboratory absorption cell measurements. They are subject to experimental errors, which, in the case of low concentrations of the absorbing gas, may be severe. Considerable extrapolation over temperature, pressure, and path length is required before application to nonhomogeneous atmospheric slant paths can be made.

(b) By theoretical calculations using band models. These are generally unsatisfactory, for reasons discussed in detail in a later section. They cannot readily be applied to certain sections of the absorption bands, which are of extreme importance in the upper atmosphere.

An alternate procedure is direct integration with respect to frequency across the absorption band, a procedure that has become increasingly attractive with the advent of modern digital computing techniques. The first serious attempt to use the method was in 1961 by Hitschfeld and Houghton,[1] who integrated over a small portion of the 9.6-μ ozone band. More recently, Gates *et al.*[2,3] used the same technique for the 1.8-μ water vapor band, and Shaw and Houghton[4] for the 4.7-μ CO band. The same general approach has been used in the calculations described in this paper, but it has been extended to include atmospheric slant paths.

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Homogeneous Paths

Line Shapes

Before atmospheric absorption calculations are performed, it is extremely important to decide on the appropriate line shape. The theory governing the shapes and half-widths is difficult to apply; furthermore, experimental work is hampered by such factors as the overlapping of lines, the difficulty in obtaining suitable high-resolution instruments, and the effect of instrument aperture functions on the spectra. However, there is good evidence[5] to support the use of the Lorentz line shape where pressure-broadening is the dominant feature and the mixed Doppler-Lorentz line shape at lower pressures.

The Lorentz Line Shape

This shape has a very simple form and has the great advantage that it is easy to deal with analytically. The absorption coefficient at frequency ν, for a single line strength S, centered at frequency ν_0, is given by

$$k_\nu = \frac{S}{\pi} \frac{\alpha_L}{(\nu - \nu_0)^2 + \alpha_L^2}, \qquad (1)$$

where α_L is the Lorentz half-width (i.e., half the total line-width at half-intensity) at temperature T and pressure p. The dependence of α_L upon these parameters is given by

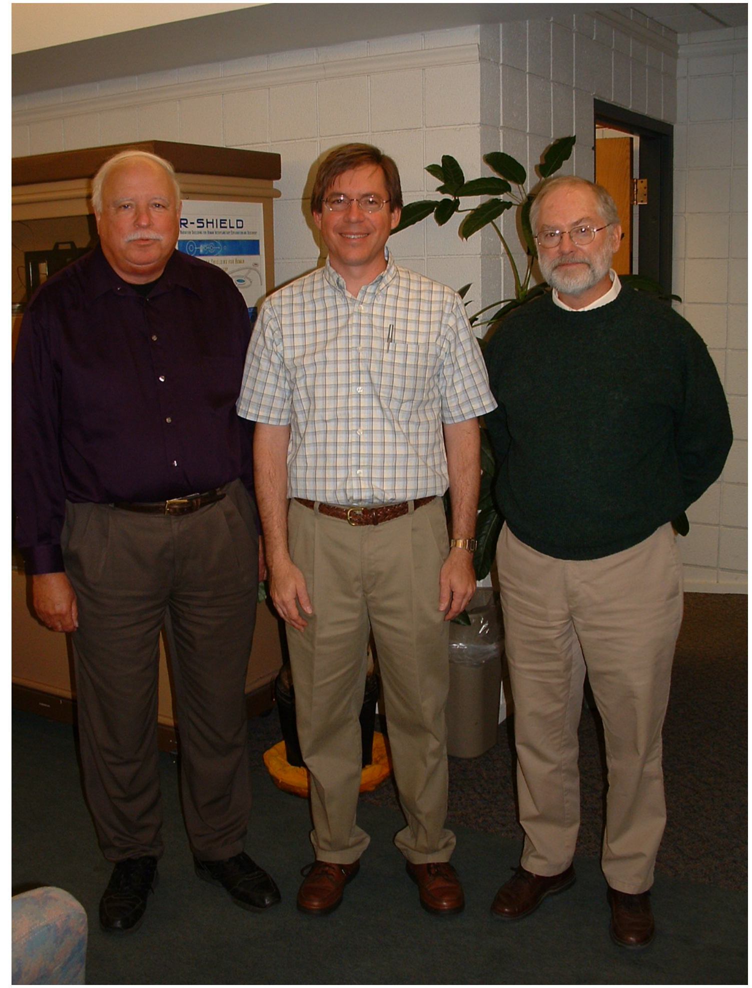
U. Michigan Recognition of Roland Drayson

"The line-by-line method of calculating atmospheric transmittance developed by Professor Drayson is used today as a standard in the field. His analyses of data taken by the Limb Infrared Monitoring of the Stratosphere (LIMS) satellite represent the first application of the detailed spectroscopy of atmospheric gases, carbon dioxide in particular"

"The Regents salute this distinguished scholar by naming S. Roland Drayson professor emeritus of atmospheric, oceanic and space sciences"

From the proclamation of the University of Michigan Board of Regents on the occasion of Professor Drayson's retirement

Professor S. Roland Drayson



2004

Motivation

- Radiative forcing (RF) by CO_2 is the leading contribution to anthropogenic climate change
- Uncertainties in CO_2 RF impact scientific and policy assessments
- Goals of this work:
  - Assess uncertainty in RF associated with infrared spectroscopy of CO_2
  - Of particular interest is line mixing and line shape function
  - Refute recent assertions that RF is greatly overestimated due to inappropriate use of Voigt lineshape (Happer, 2014)

- ***Result***: RF spectroscopic uncertainty is < 1% and the foundation of climate change modeling is robust!

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Why has global warming paused?*

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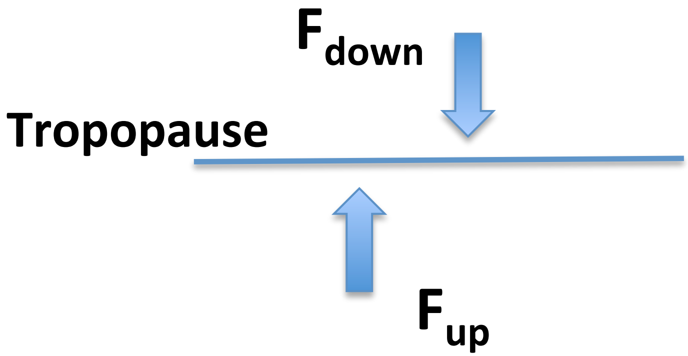
Definition of Radiative Forcing (RF) - 1

- RF is the ***change*** in the ***net*** radiative flux F_{net} at the ***tropopause***

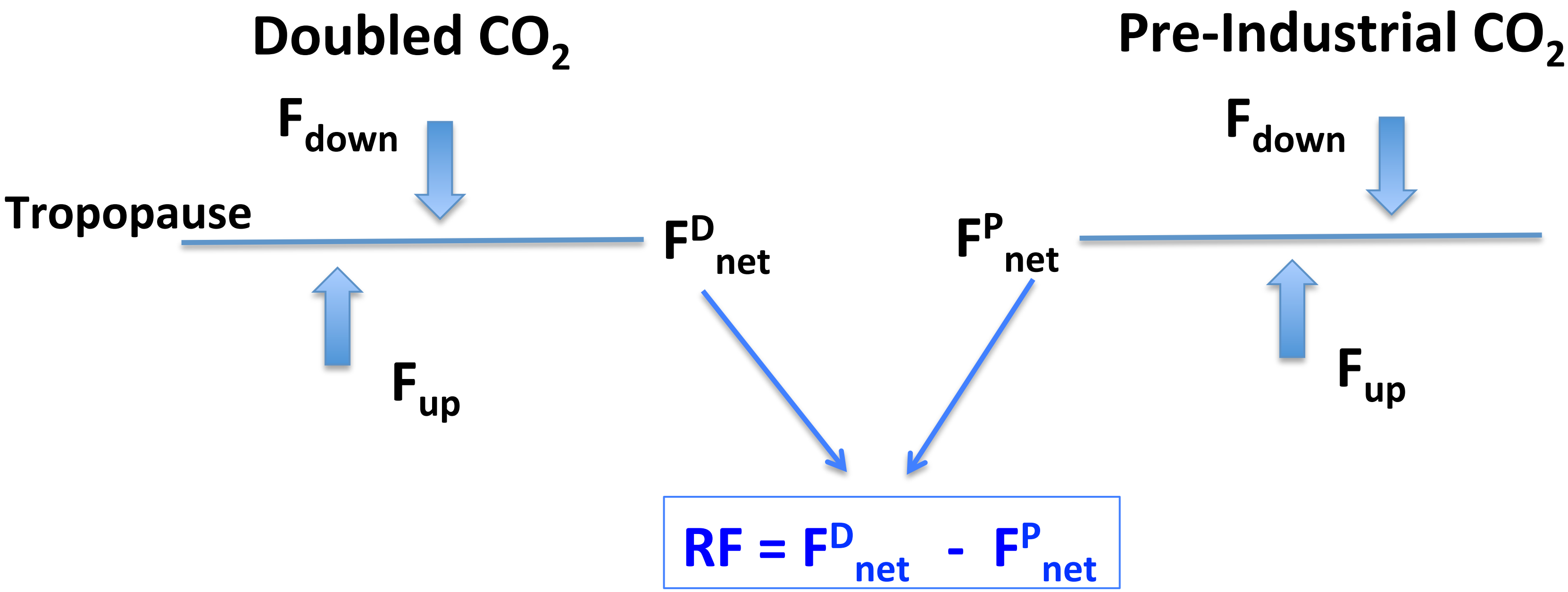
- ***Net flux*** is defined as F_{down} *minus* F_{up}



$F_{net} = F_{down} - F_{up}$

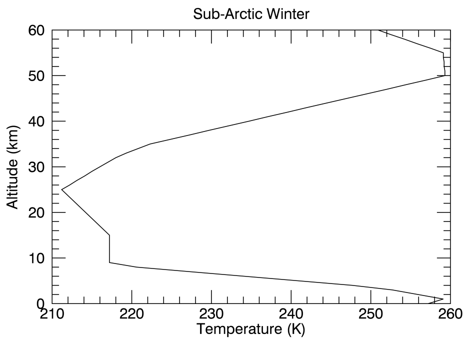
Definition of Radiative Forcing (RF) - 2

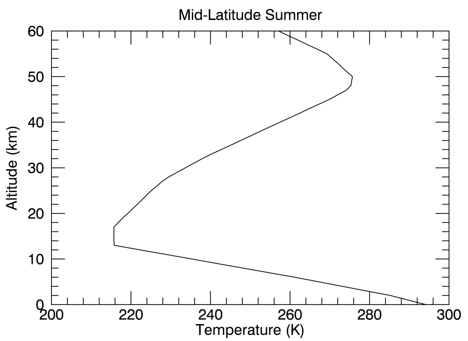
- RF is ***difference*** in net flux for two different CO_2 burdens
- Doubled CO_2 from pre-industrial CO_2 (560 ppm and 280 ppm)



Calculating Radiative Forcing

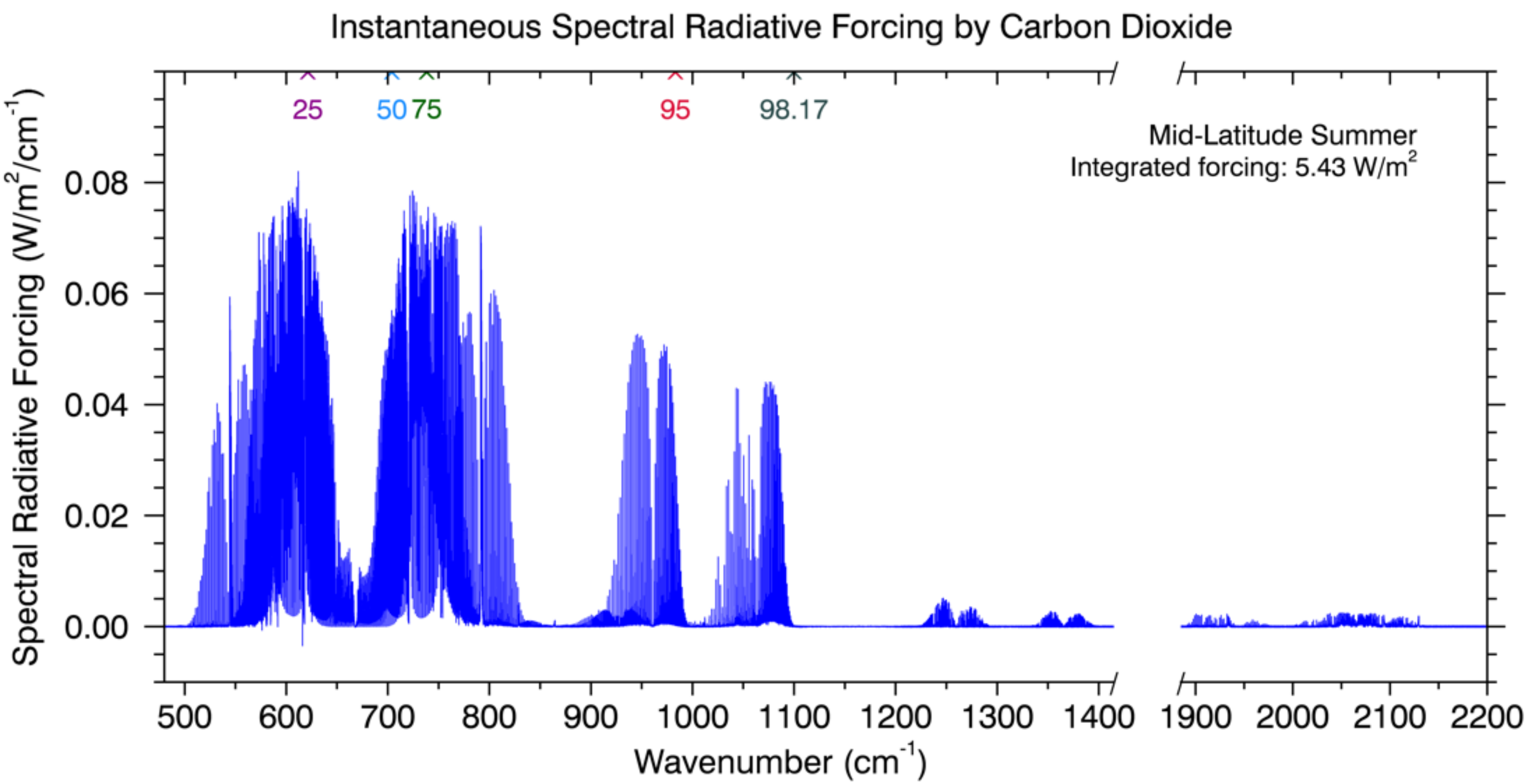
- Use LBLRTM model, v12.2, and v3.2 spectral line database
  - Compute radiances and use 3 point Gaussian integration for fluxes
- Compute <u>*instantaneous*</u> radiative forcing
- Five “McClatchey” standard atmospheres
  - Mid-latitude summer, Mid-latitude winter
  - Sub-arctic summer; Sub-arctic winter;
  - Tropical annual
- Include CH_4, CO, and N_2O as well-mixed gases
- H_2O and O_3 as per the profile
- Clear sky conditions





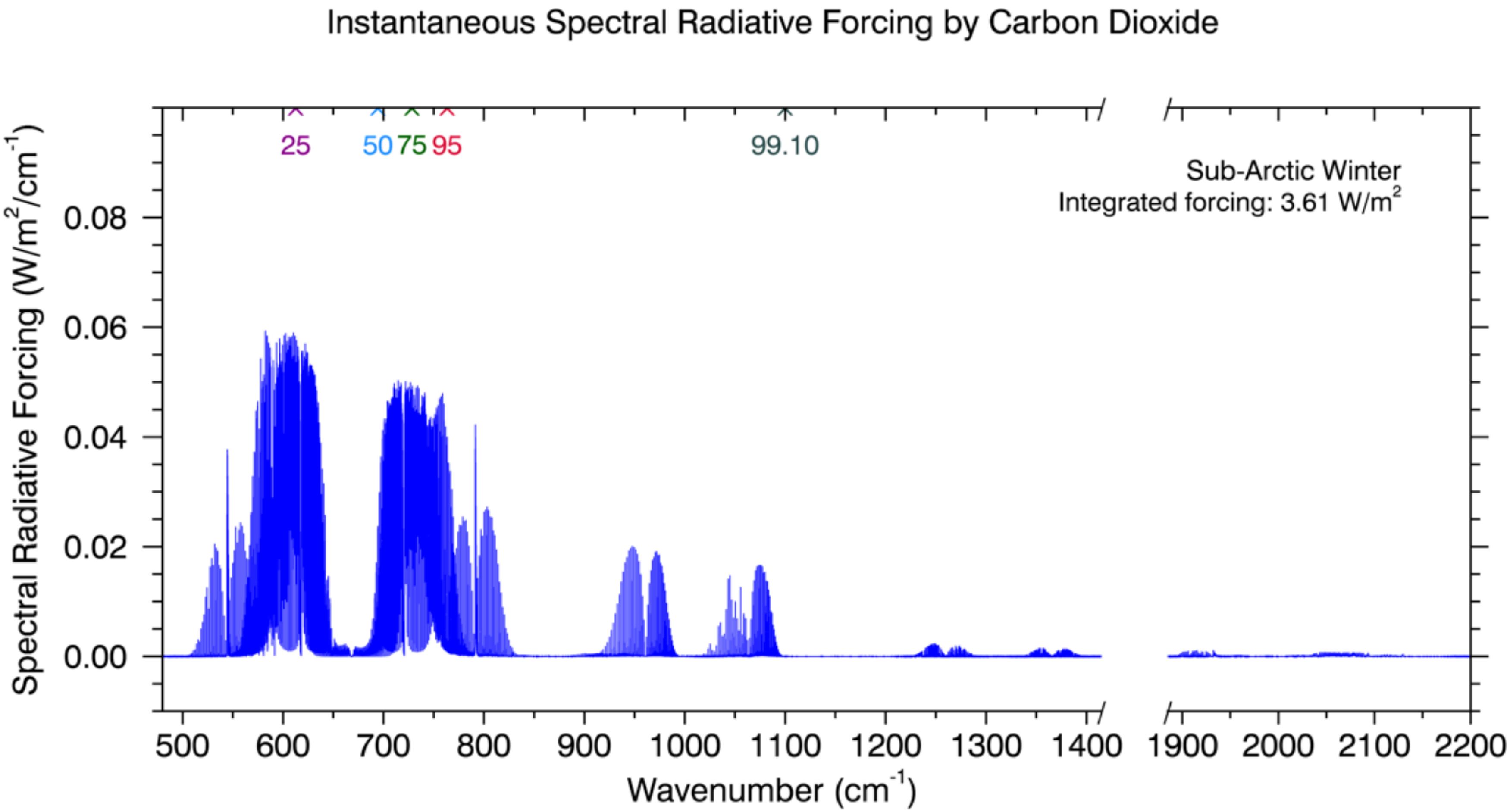
- *Compute the <u>spectrum</u> of radiative forcing*

The Spectrum of Radiative Forcing for 2 x CO_2



Baseline Case, Mid-Latitude Summer

The Spectrum of Radiative Forcing for 2 x CO_2



Baseline Case, Sub-Arctic Winter

Spectroscopic Uncertainty - Methodology

- **Only uncertainty in the transmittance function is considered:**

$$T_\nu(z,z') = \exp\left(-\sum_i S_i(\Theta) g_i(\nu - \nu_0) \frac{u(z,z')}{\mu} \right)$$

  - **Line shape function $g(\nu - \nu_o)$**
  - **Line strength $S_i(\Theta)$**
  - **Air-broadened halfwidth (in lineshape function)**

- **Perturb these parameters to assess uncertainty in spectroscopy; compute difference with "baseline" cases**

Uncertainties in Spectroscopy: Line Shape/Mixing

- Classically, the Voigt lineshape is used to represent the effects of collision/Lorentz and Doppler broadening
- Wings of many IR-active species are sub- or super-Lorentzian
- The 15-μm bands of CO_2, lines are densely packed and cannot be considered isolated during collisions
- Line mixing occurs, resulting in additional absorption near line centers and sub-Lorentzian behavior in the line wings
  - There is a 50-year history of modeling sub-Lorentzian wings in CO_2
- LBLRTM adopts line mixing from Niro et al. (2005)
  - Replaces classic line shape function in transmittance calculation

Approach to Computing Spectroscopic Uncertainty

- **Uncertainty determined by perturbation analysis**

- **Compute RF for "Baseline" case**

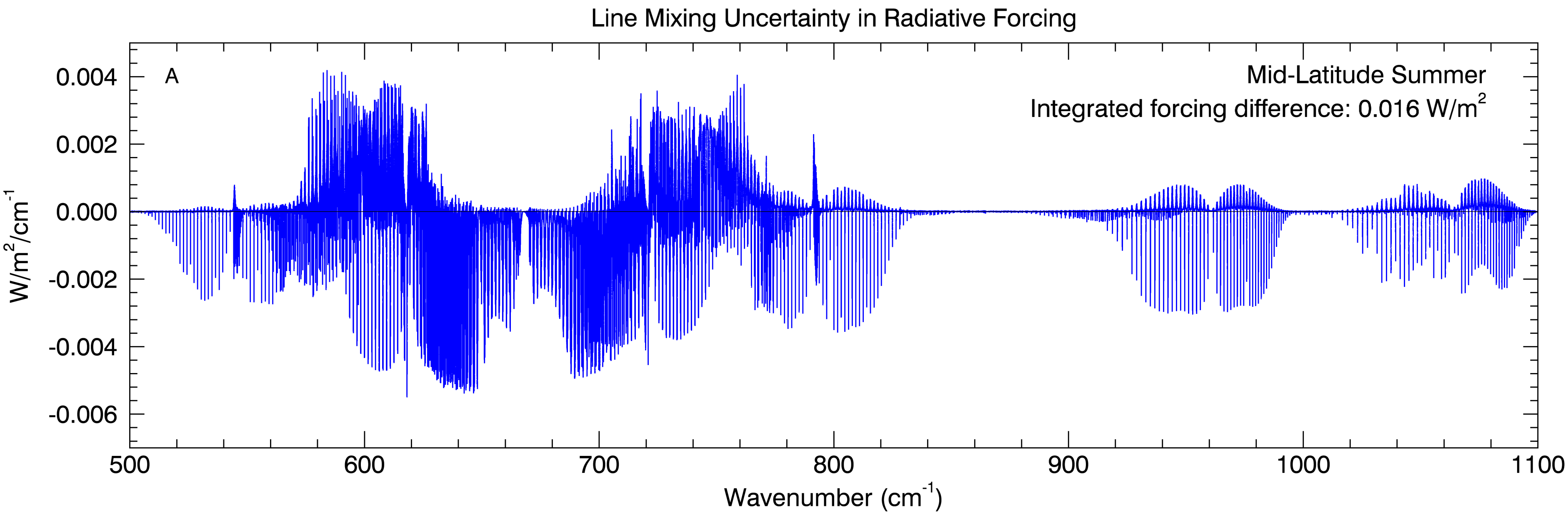
- **Compute RF for "Perturbed" case**
  - Increase CO_2 line mixing by 20%
  - Perturb CO_2 line strengths by assigned uncertainty on AER v3.2 database
  - Perturb CO_2 line halfwidths by assigned uncertainty on AER v3.2 database

- **Compute Uncertainty = Perturbed RF *minus* Baseline RF**
  - Spectra, and spectrally integrated differences
  - RSS all uncertainties to get total uncertainty

Uncertainty in Radiative Forcing due to CO_2 Line Mixing

RF (increased line mixing) – RF (baseline)

Mid-Latitude Summer Atmosphere



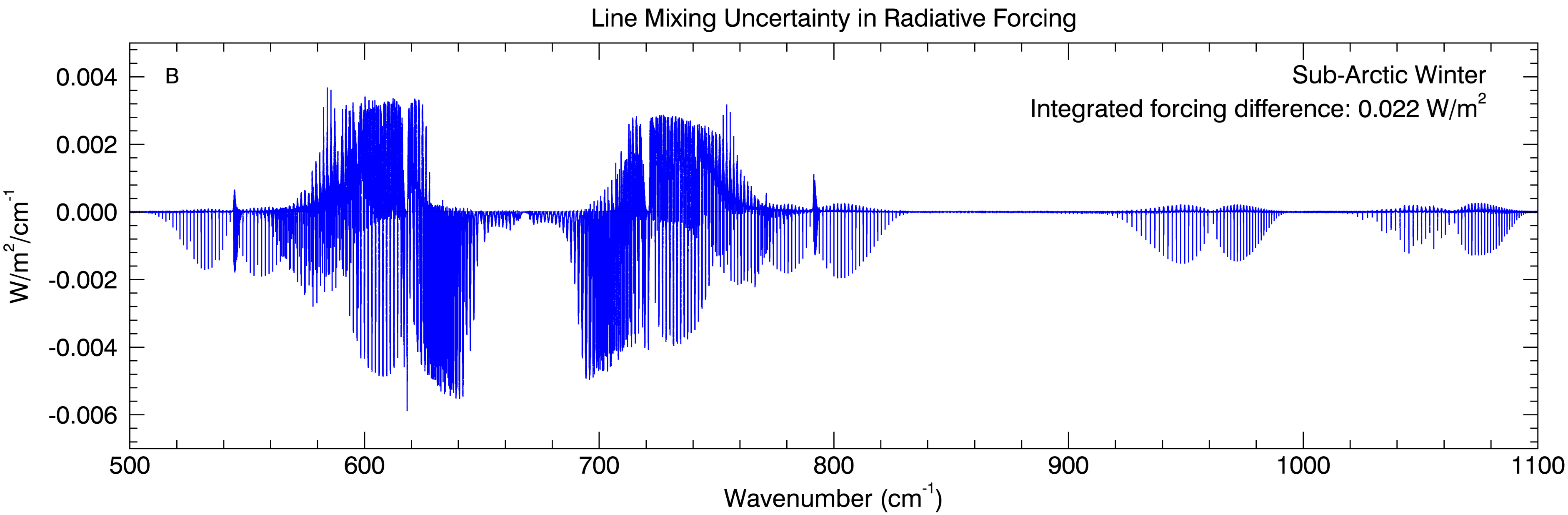
Significant but compensating spectral structure

RF difference is 0.016 W/m^2 or 0.3% of RF baseline

Uncertainty in Radiative Forcing due to CO_2 Line Mixing

RF (increased line mixing) – RF (baseline)

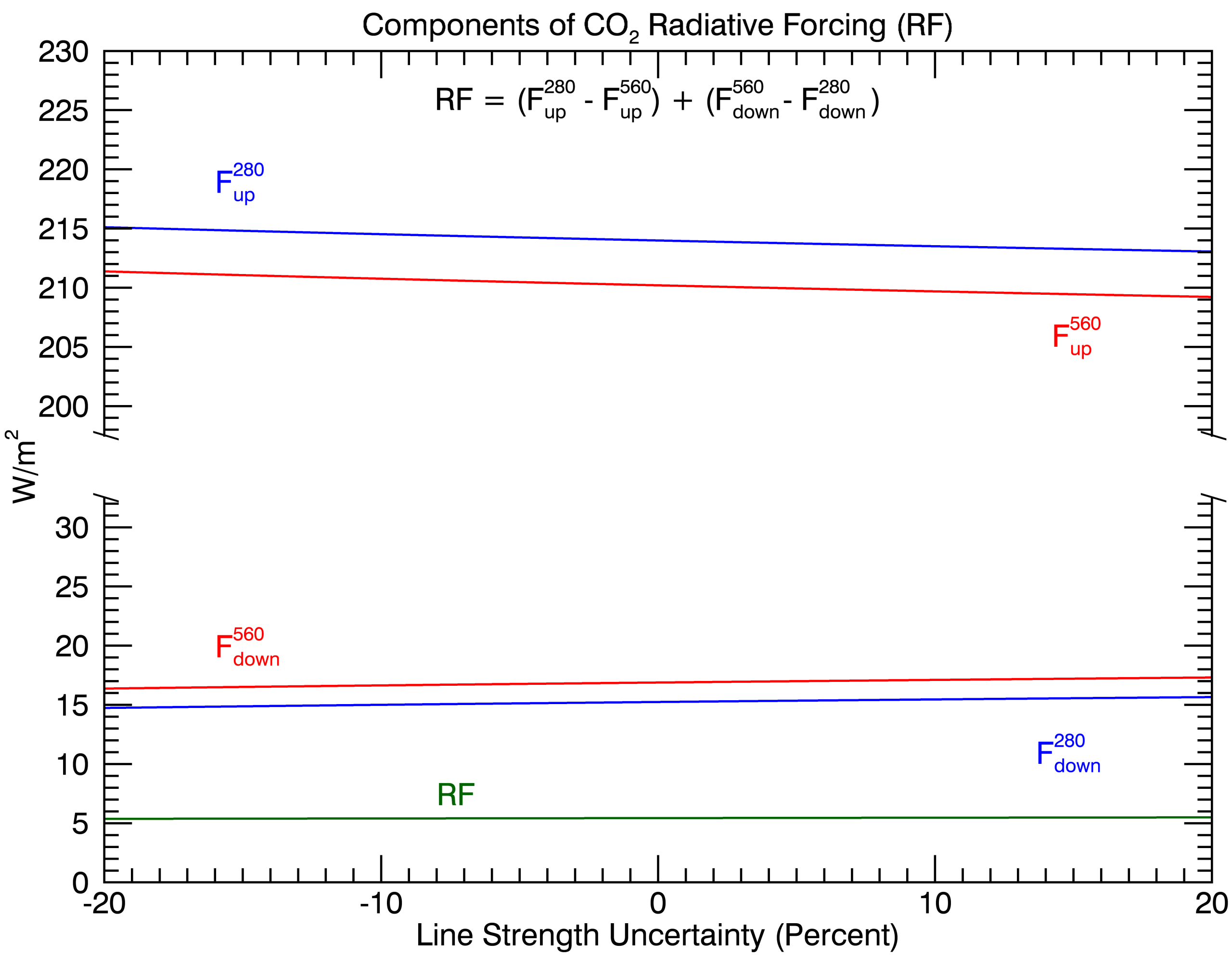
Sub-Arctic Winter Atmosphere



Significant but compensating spectral structure

RF difference is 0.022 W/m^2 or 0.6% of RF baseline

CO_2 Radiative Forcing Uncertainty due to Line Strength



Virtually no change in RF with up to 20% increase/decrease in S

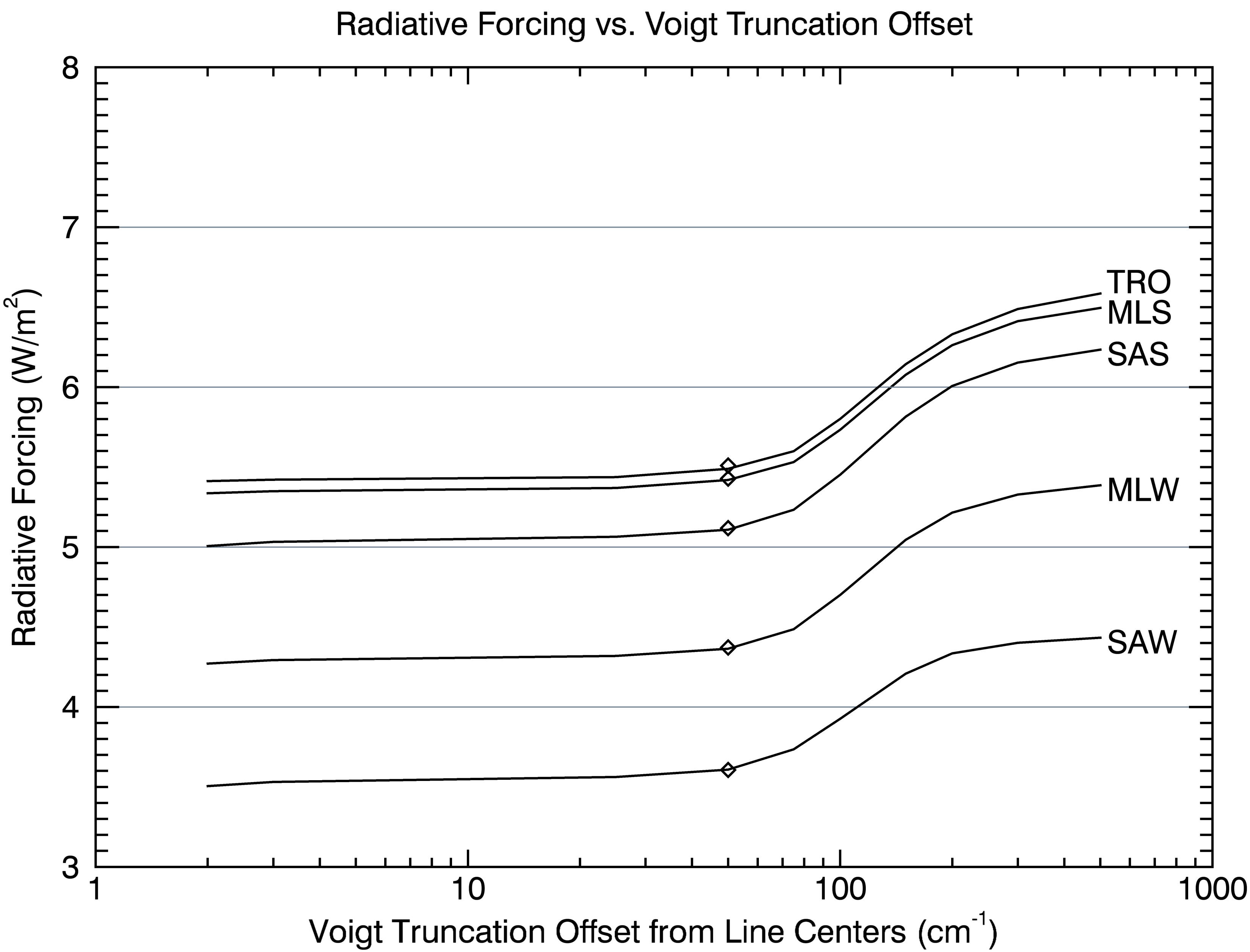
Summary of Spectroscopic Uncertainty in CO_2 RF

Atmosphere	Line Shape Uncertainty W/m^2	Line Strength Uncertainty W/m^2	Halfwidth Uncertainty W/m^2	RSS W/m^2	Error as % of Baseline RF
Mid-Latitude Summer	0.016	0.015	0.005	0.022	0.41
Mid-Latitude Winter	0.016	0.010	0.004	0.019	0.44
Sub-Arctic Summer	0.023	0.009	0.007	0.028	0.55
Sub-Arctic Winter	0.022	0.015	0.006	0.025	0.68
Tropical	0.010	0.014	0.002	0.017	0.51

Spectroscopic Uncertainty in RF is < 0.7% of Forcing in a Variety of Atmospheres

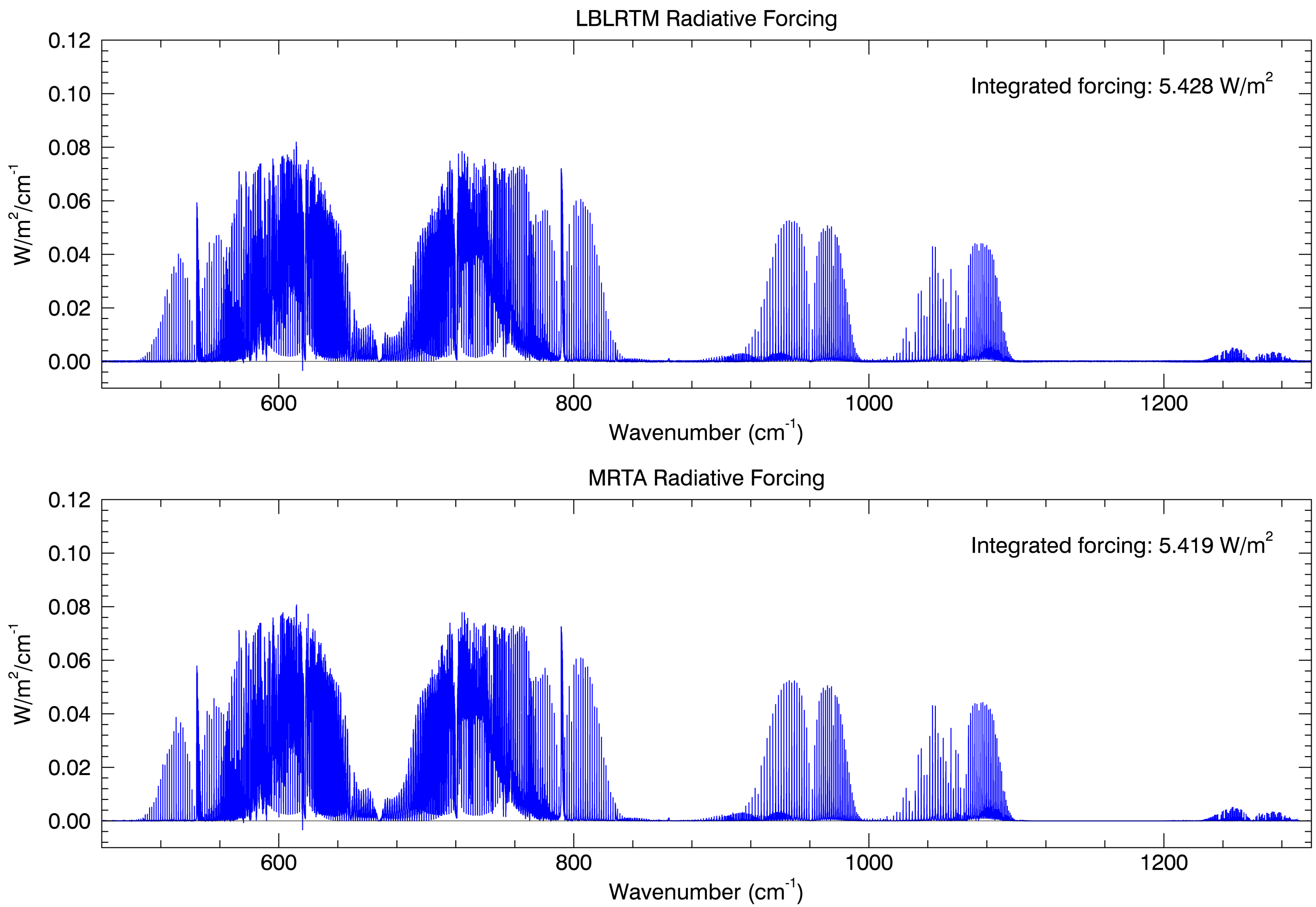
Utility of the Voigt Profile

- Examine the utility of the Voigt profile by computing the RF and truncating the Voigt wings at different distances from line center
- At 50 cm-1 truncation, Voigt-only agrees almost identically with LBLRTM with full line mixing
- At larger truncations, Voigt-only begins to exceed LBLRTM by up to 25% at 500 cm-1 truncation

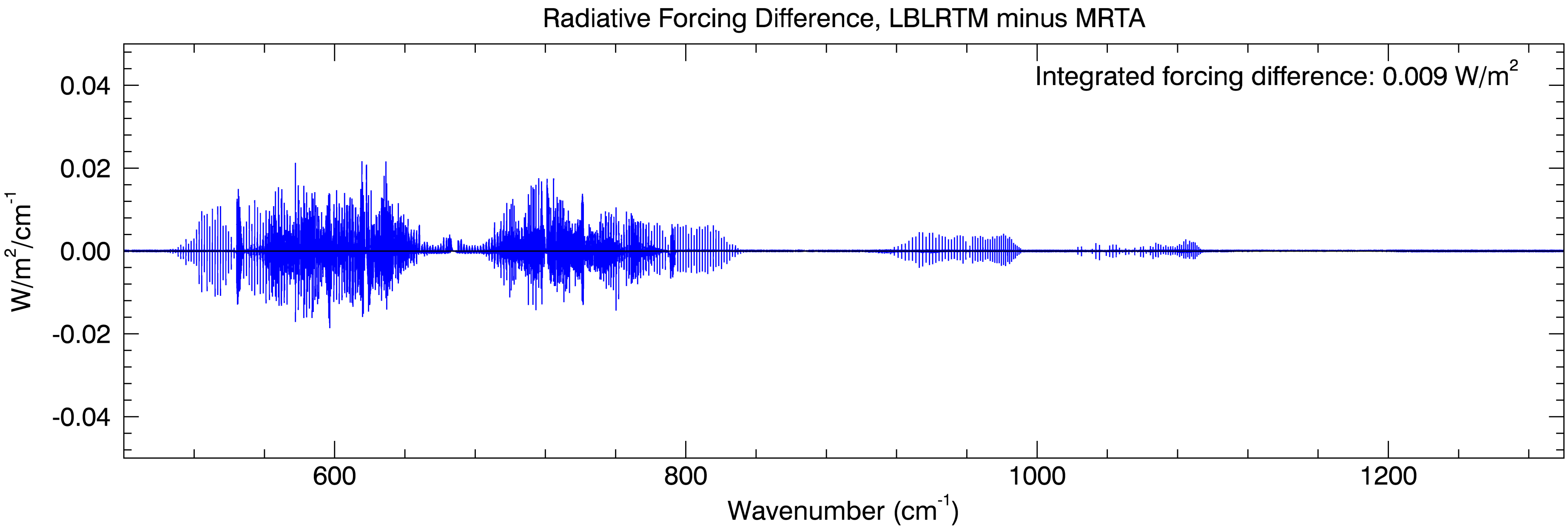


Radiative Forcing vs. Voigt Truncation Offset
Radiative Forcing (W/m2)
Voigt Truncation Offset from Line Centers (cm-1)
TRO
MLS
SAS
MLW
SAW
3
4
5
6
7
8
1
10
100
1000

Comparison of LBLRTM w/Full Line Mixing and Voigt-only with 50 cm^{-1} Truncation



Difference: LBLRTM minus Voigt-only



Summary

- We have examined uncertainty in radiative forcing by CO_2 associated with spectroscopic parameters
- Combined uncertainty of line shape, line strength, and halfwidth is < 0.7% for a variety of standard atmospheres
- Line mixing is rigorously included in state of the art line-by-line models and in rapid codes used in climate models
  - Extending Voigt "to infinity" is never done in contemporary models
- Foundation of climate modeling is robust with regards to uncertainties in spectroscopy

Status

- Journal article describing these results accepted in GRL 22 April 2016

- Accepted manuscript available online now

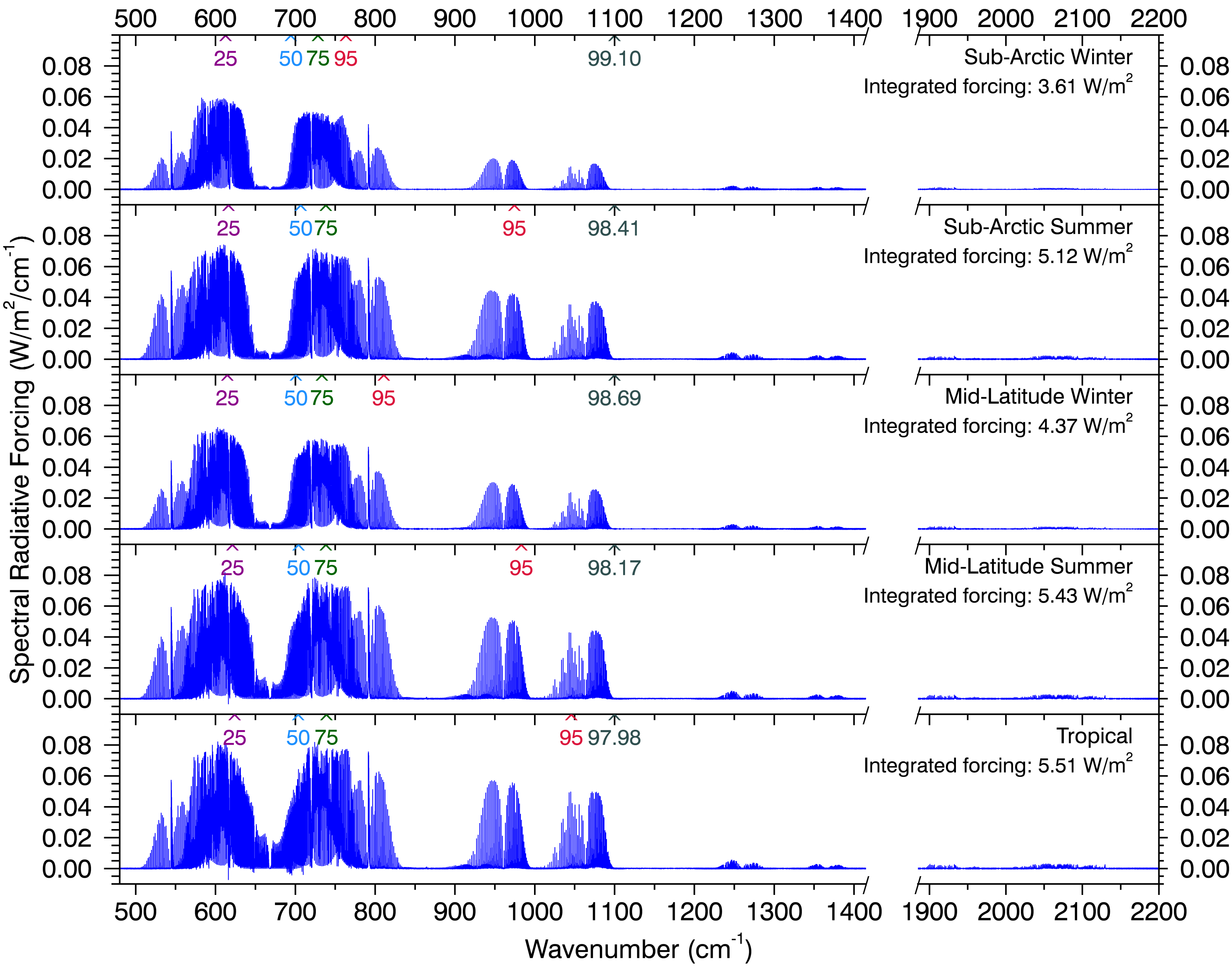
- Actual journal article available online shortly

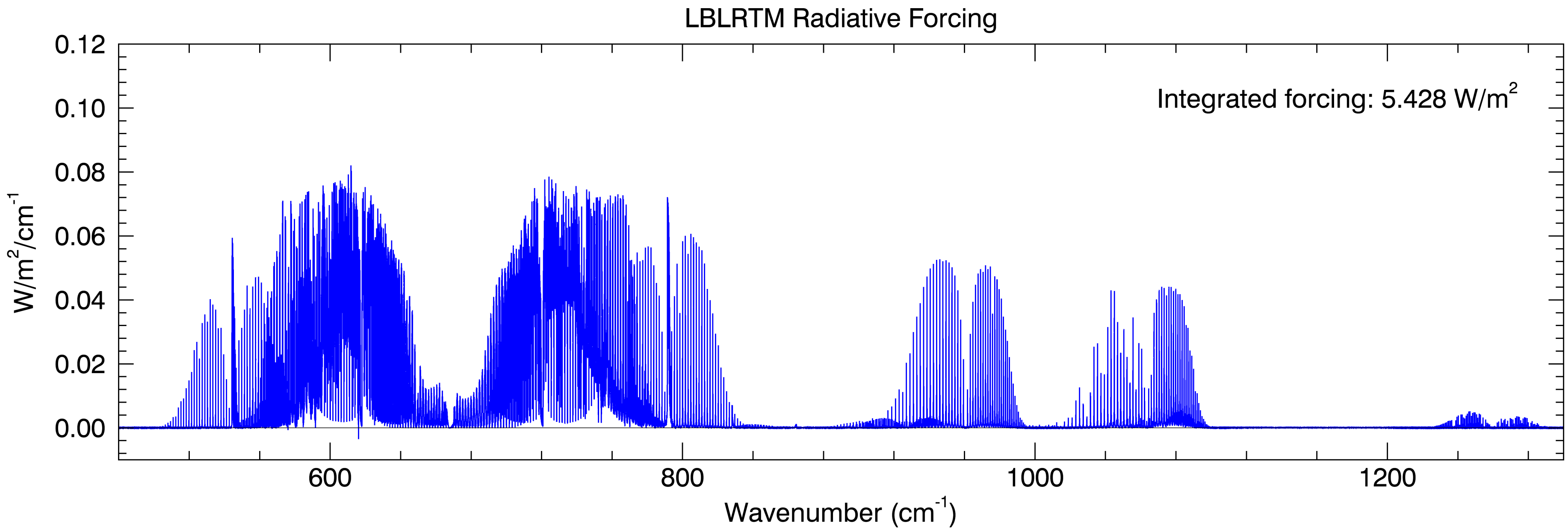
Backup Slides

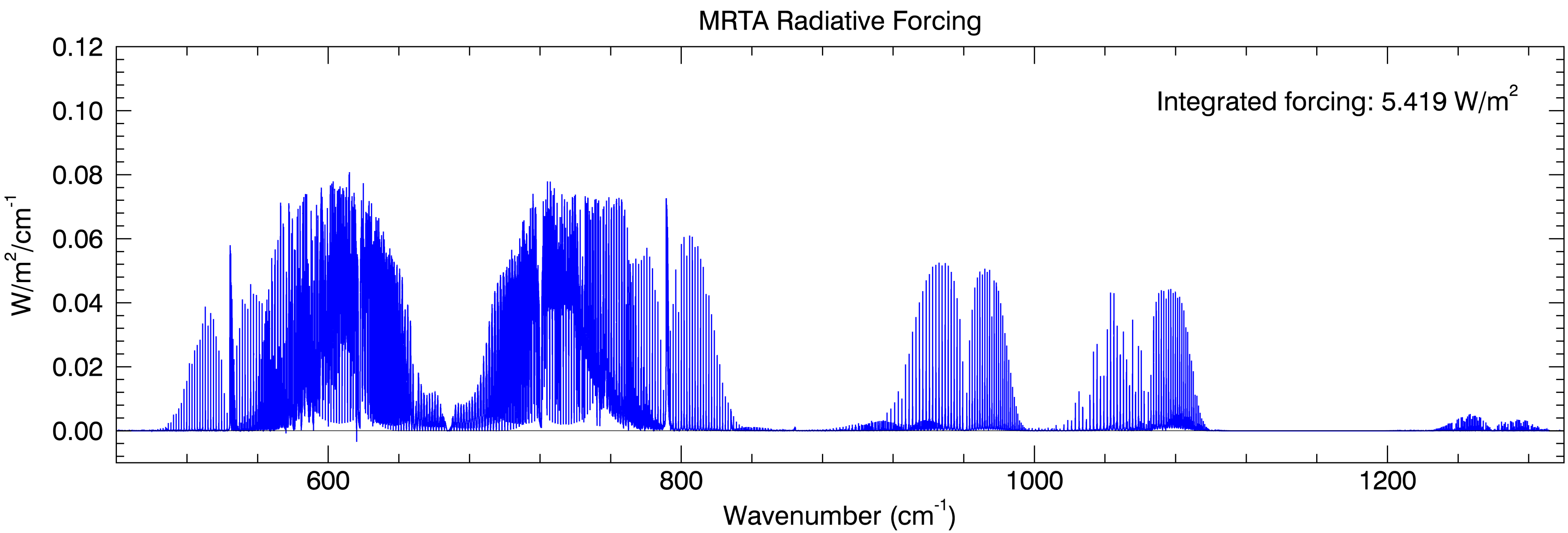
Summary of Spectroscopic Uncertainty in RF

Atmosphere	RF LBLRTM Baseline W/m^2	RF LBLRTM 20% Pert. W/m^2	Uncertainty Line Shape/Line Mixing W/m^2	Uncertainty Line Strength W/m^2	Uncertainty Half Width W/m^2	RSS W/m^2	RSS as Percent of baseline LBLRTM Forcing
MLS	5.428	5.444	0.016	0.015	0.005	0.022	0.41
MLW	4.372	4.388	0.016	0.010	0.004	0.019	0.44
SAS	5.118	5.141	0.023	0.015	0.007	0.028	0.55
SAW	3.606	3.628	0.022	0.009	0.006	0.025	0.68
TRO	5.509	5.519	0.010	0.014	0.002	0.017	0.31

Instantaneous Spectral Radiative Forcing by Carbon Dioxide

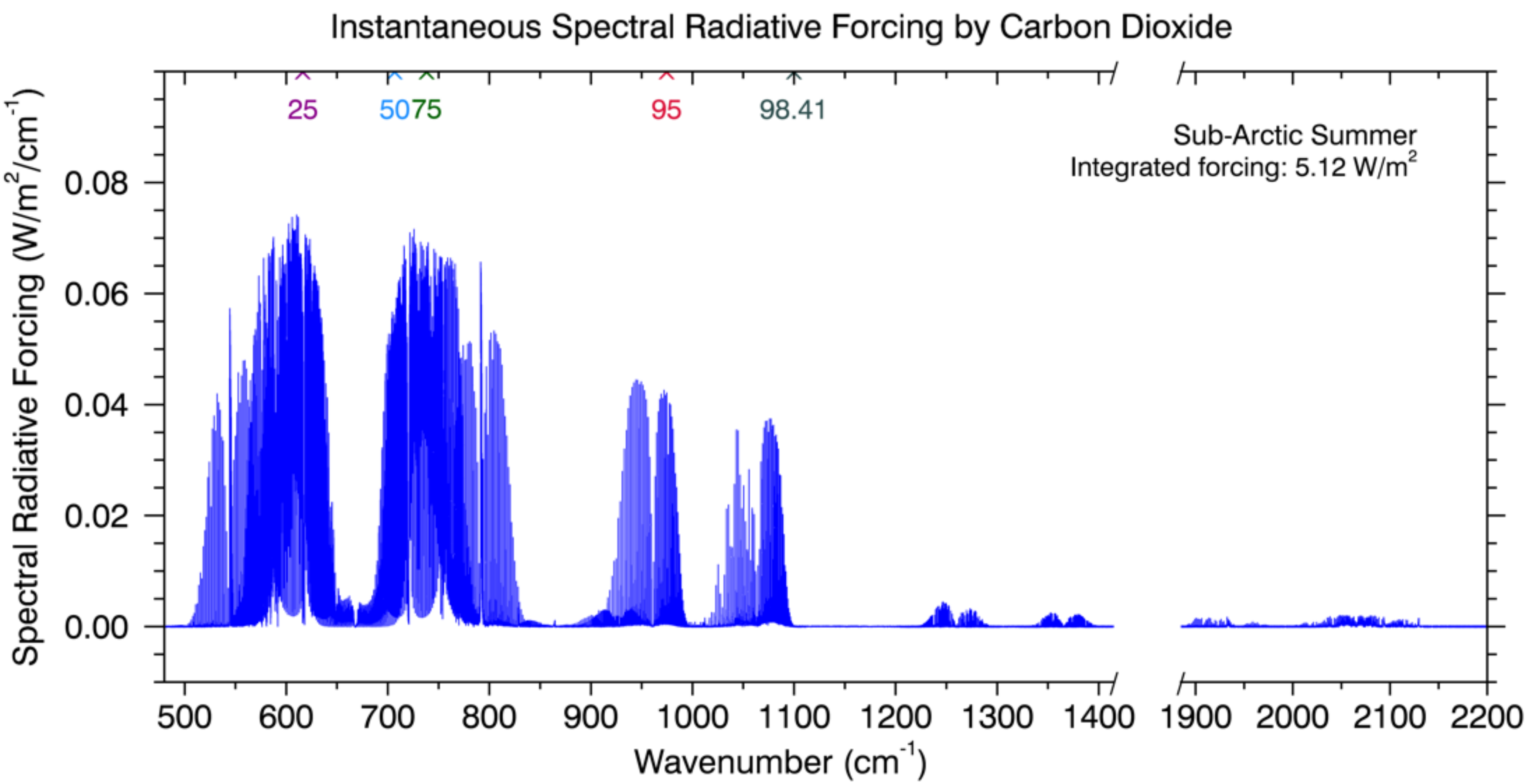



LBLRTM Radiative Forcing
Integrated forcing: 5.428 W/m2
0.12
0.10
0.08
0.06
0.04
0.02
0.00
W/m2/cm-1
600
800
1000
1200
Wavenumber (cm-1)

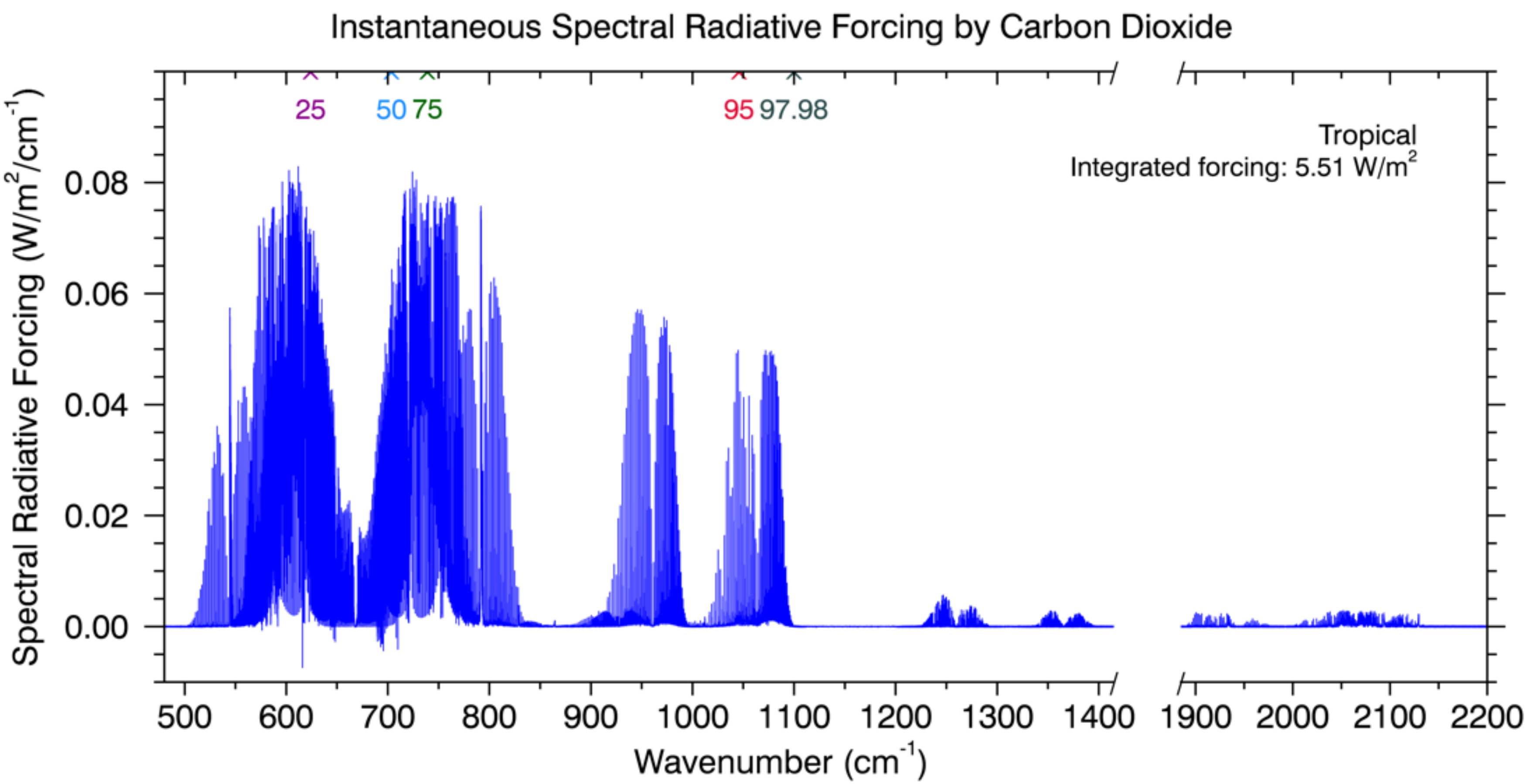


MRTA Radiative Forcing
Integrated forcing: 5.419 W/m^2
0.12
0.10
0.08
0.06
0.04
0.02
0.00
W/m^2/cm^{-1}
600
800
1000
1200
Wavenumber (cm^{-1})

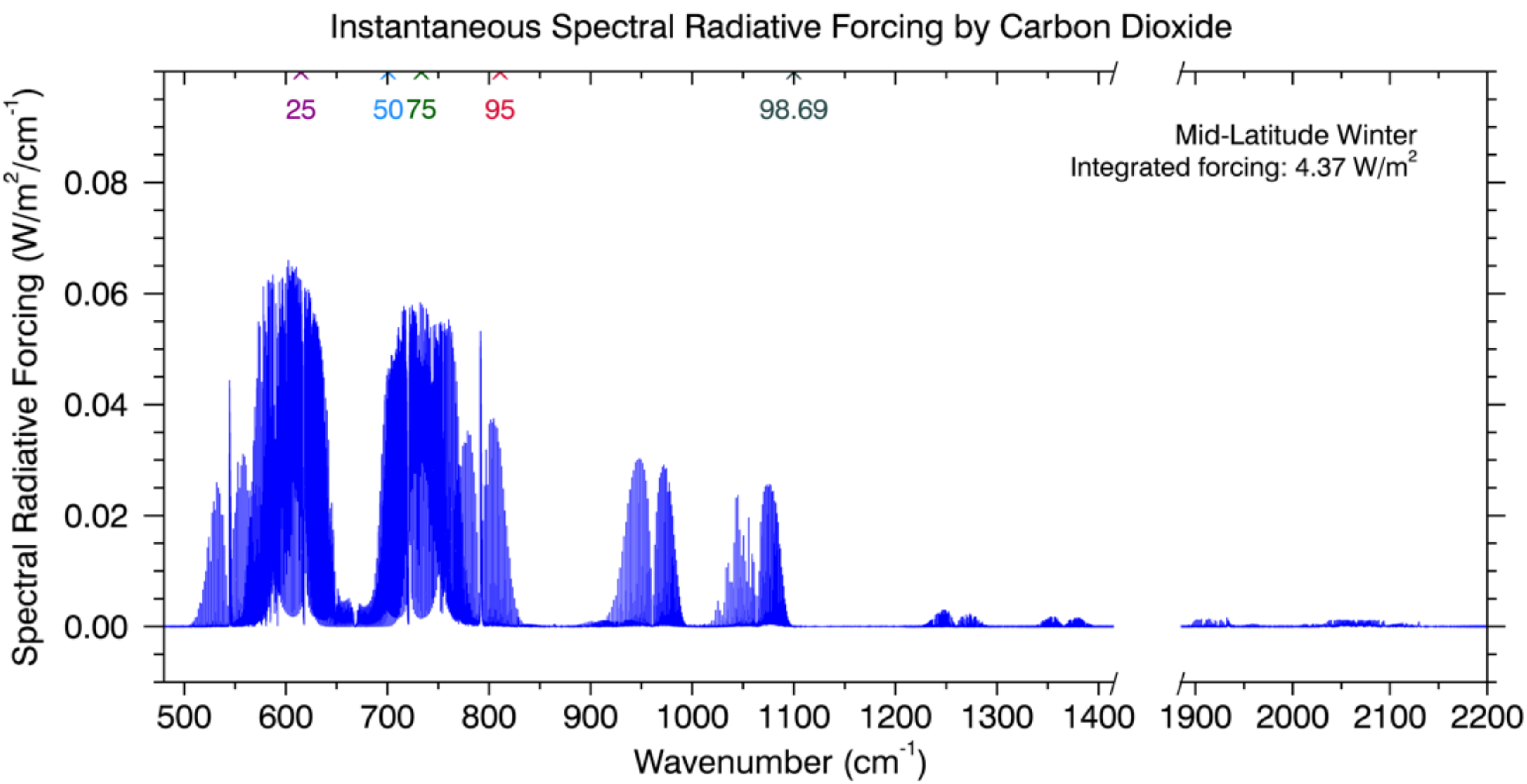
The Spectrum of Radiative Forcing

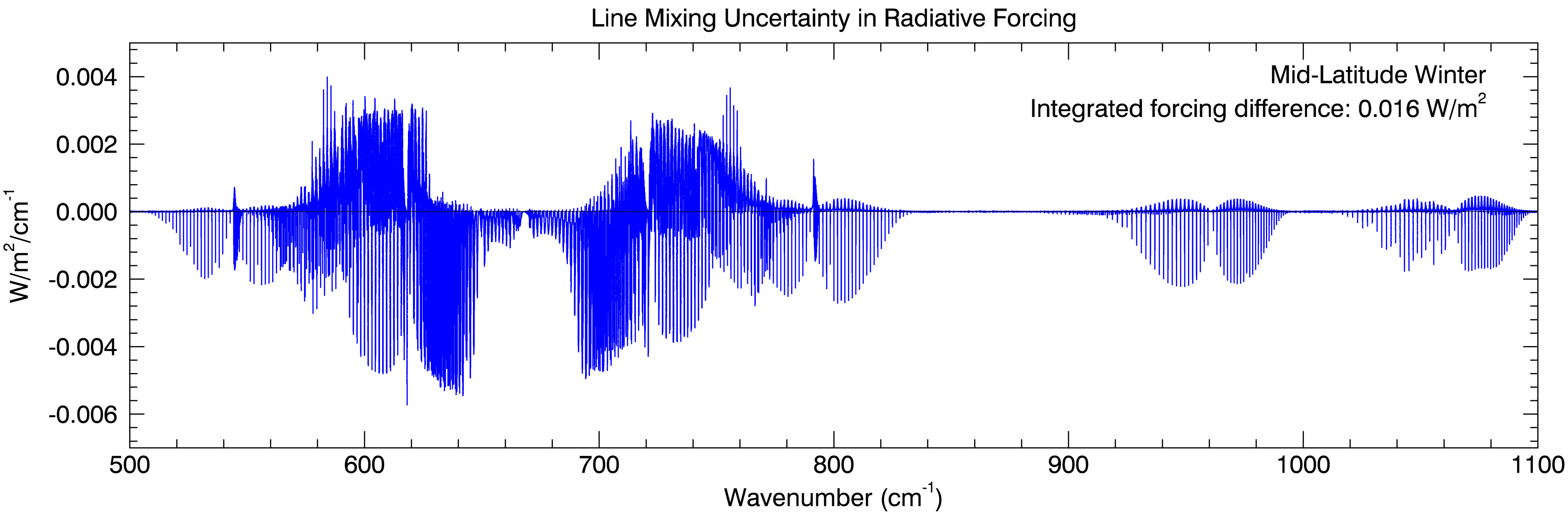


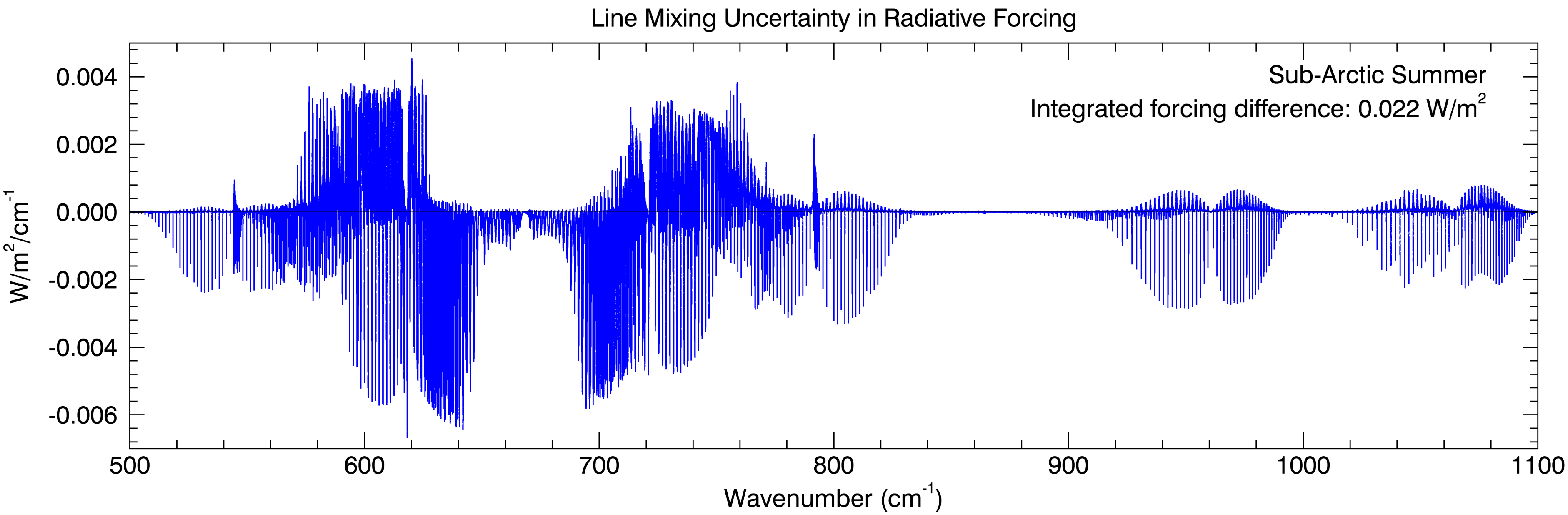
The Spectrum of Radiative Forcing

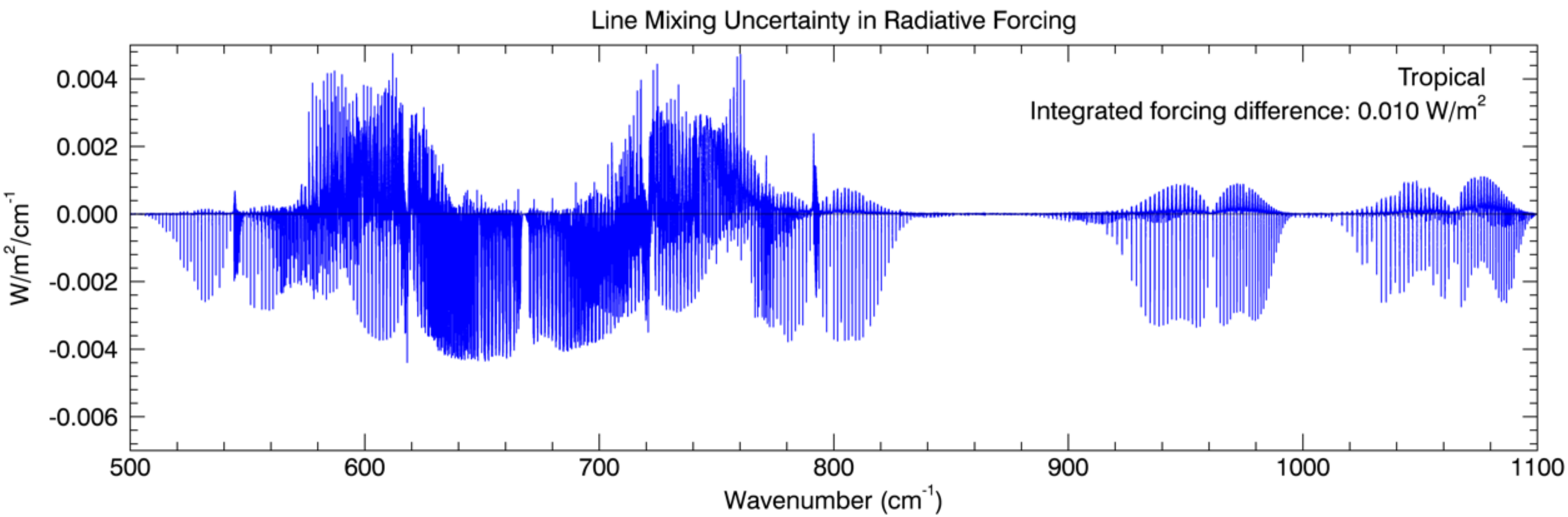


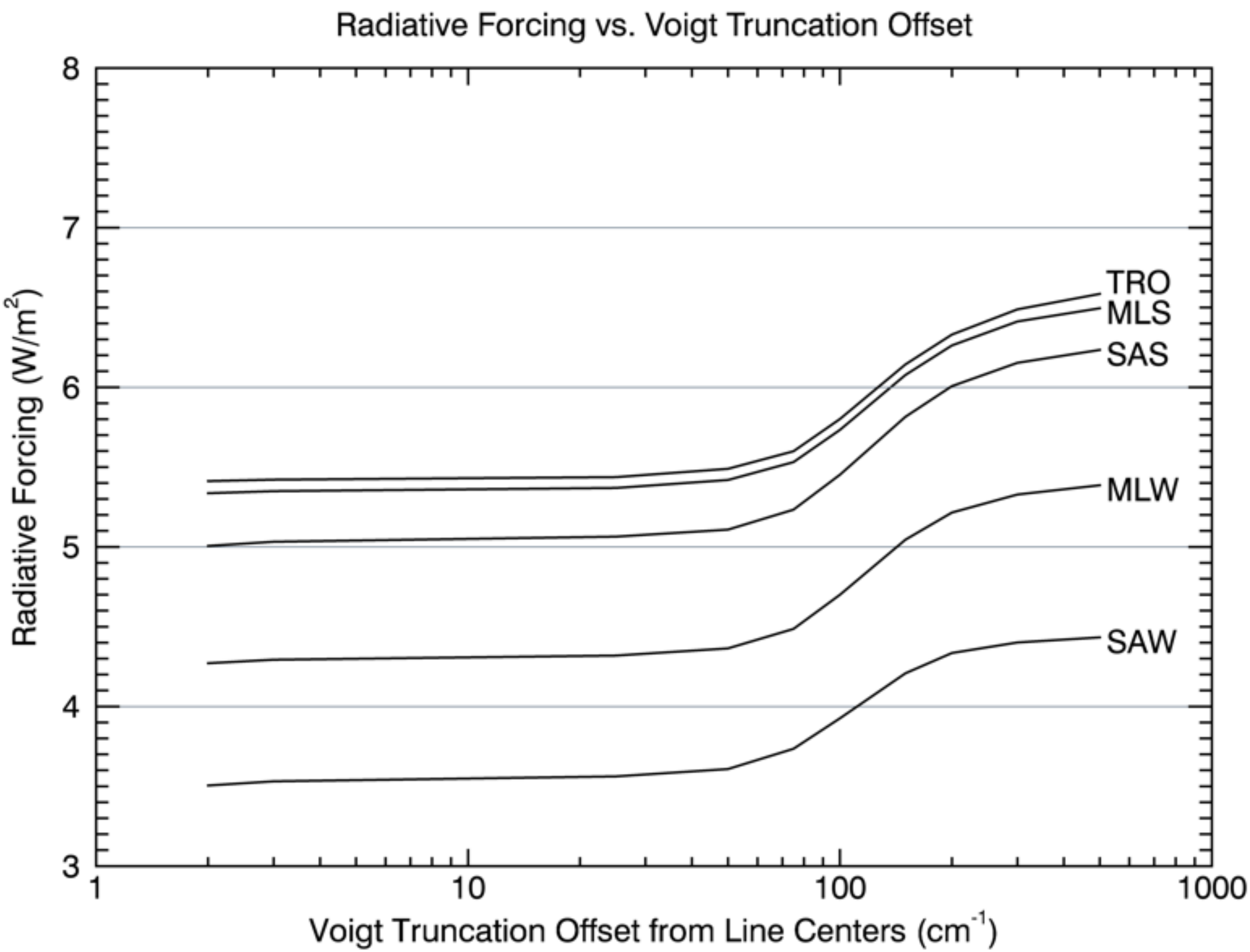
The Spectrum of Radiative Forcing




Line Mixing Uncertainty in Radiative Forcing
Mid-Latitude Winter
Integrated forcing difference: 0.016 W/m2
0.004
0.002
0.000
-0.002
-0.004
-0.006
W/m2/cm-1
500
600
700
800
900
1000
1100
Wavenumber (cm-1)


Line Mixing Uncertainty in Radiative Forcing
Sub-Arctic Summer
Integrated forcing difference: 0.022 W/m2
0.004
0.002
0.000
-0.002
-0.004
-0.006
W/m2/cm-1
500
600
700
800
900
1000
1100
Wavenumber (cm-1)

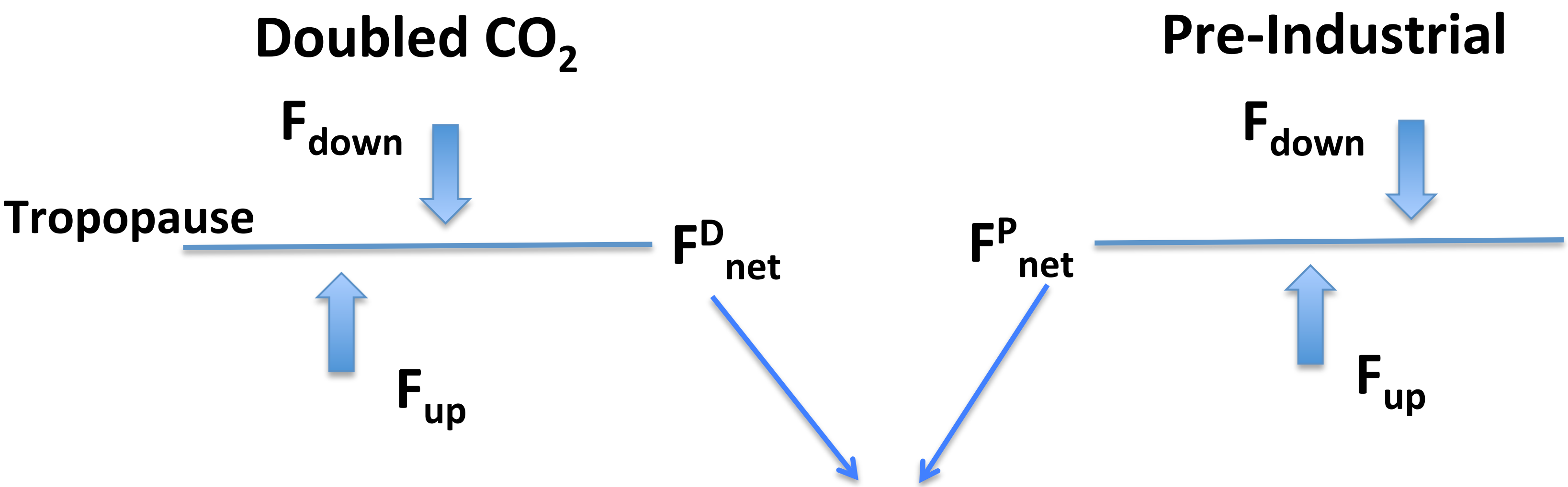

Line Mixing Uncertainty in Radiative Forcing
Tropical
Integrated forcing difference: 0.010 W/m^2
W/m^2/cm^{-1}
0.004
0.002
0.000
-0.002
-0.004
-0.006
500
600
700
800
900
1000
1100
Wavenumber (cm^{-1})



Radiative Forcing vs. Voigt Truncation Offset
Radiative Forcing (W/m^2)
Voigt Truncation Offset from Line Centers (cm^-1)
8
7
6
5
4
3
1
10
100
1000
TRO
MLS
SAS
MLW
SAW

Definition of Radiative Forcing (RF)

- RF is the ***change*** in the ***net*** radiative flux at the ***tropopause***
- ***Net flux*** is defined as F(down) minus F(up)
- ***Change*** in net flux is difference for two different CO_2 burdens, typically doubled from pre-industrial (PI) minus PI



$$\boxed{RF = F^D{}_{net} - F^P{}_{net}}$$

Methodology

- **Use LBLRTM v12.2 to evaluate up and down radiances/fluxes**

$$I_\nu(\mu, z) = B_\nu(\Theta_s) T_\nu(z, 0) + \int_0^z B_\nu(\Theta(z')) \frac{\partial T_\nu(z, z')}{\partial z'} dz'$$

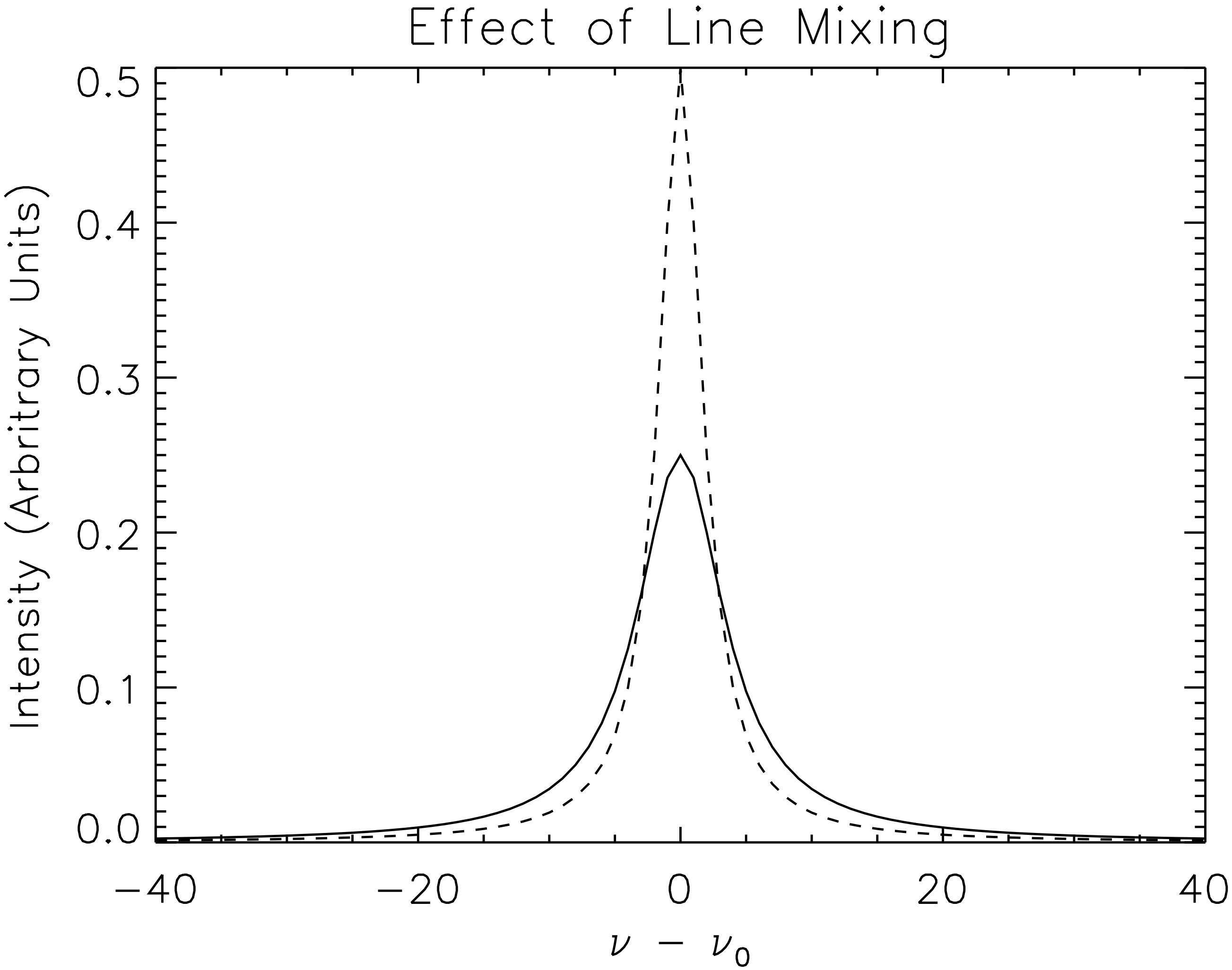
$$I_\nu(-\mu, z) = -\int_z^\infty B_\nu(\Theta(z')) \frac{\partial T_\nu(z, z')}{\partial z'} dz'$$

- **The key to our study is the transmittance function:**

$$T_\nu(z, z') = \exp\left(-\sum_i S_i(\Theta) g_i(\nu - \nu_0) \frac{u(z, z')}{\mu} \right)$$

- **Compute instantaneous forcing, no change in temperature**

- **Vary $g(\nu - \nu_o)$, S_i, and α_L to assess *<u>spectroscopic</u>* uncertainty in RF**


Effect of Line Mixing
0.5
0.4
0.3
0.2
0.1
0.0
Intensity (Arbitrary Units)
−40
−20
0
20
40
ν − ν0